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(54) Title: RESIN COMPOSITION FOR BIODEGRADABLE AGRICULTURAL FILMS WITH ENHANCED WEATHERABILITY

(57) Abstract

The invention relates to a biodegradable resin composition and films and sheets made thereof with improved heat resistance and weather resistance when used as final products in outdoor applications while maintaining excellent biodegradability and compostability. The products are used in the fields of agriculture, packaging and civil engineering such as a mulch film, polytunnel, greenhouse, masking film, riverbed sheet, stretch film for pallets, and silage film.

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Resin composition for biodegradable agricultural films with enhanced weatherability

Industrial Field of The Invention

The present invention relates to a biodegradable resin composition and films and sheets made thereof with improved heat resistance and weather resistance when used as final products in outdoor applications while maintaining excellent biodegradability and compostability. The products are used in the fields of agriculture, packaging and civil engineering such as a mulch film, polytunnel, greenhouse, masking film, riverbed sheet, stretch film for pallets, and silage film.

Description of the Related Art

Biodegradable aliphatic polyester resins of the poly(hydroxyalkanoate) type such as poly(hydroxybutyrate) are known for a long time but have the drawback of poor flexibility and transparency (WO-A 9407940, WO-A 9923161). Also well known is polylactide with good transparency but poor flexibility (JP-A 07206773). Aromatic-aliphatic copolyesters (WO-A 9615173, WO-A 9615174, WO-A 9615175, WO-A 9615176) and aliphatic copolyester resins of the poly(alkylenedialkanoate) type (EP-A 569153) also have the drawback of poor transparency. Blends and compounds based on starch and polyester resins are well known in the art as biodegradable resins (WO-A 201743, WO-A 9631561) but have the drawback of poor transparency and poor resistance against water and changes of humidity. Polyesteramides are also known (EP-A 641 817) as versatile materials appropriate for manufacturing flexible, transparent and water resistant films but are still poor in weatherability.

One of the methods for preventing weeds from budding without spraying any herbicides as well as saving the labor of manually removing the weeds is to cover the rice field with a mulch film. Besides biodegradable films based on starch-containing ma-

terials with the drawback of missing water resistance a multi-sheet of paper also has been used. While the multi-sheet of paper also requires no withdrawal or disposal after use since it is biodegradable by leaving it alone after completing cultivation, thick layers have to be applied in order to maintain enough tear resistance even in wet conditions. Another drawback of paper mulch is the poor flexibility which makes it hard to apply. No agricultural films simultaneously satisfying biodegradability, flexibility, tensile strength, extension and tear strength, transparency, cloud preventive property, durability, heat and water resistance have been reported yet.

Problems to be Solved by the Invention

Accordingly, the object of the present invention is to provide a composition for manufacturing films and sheets having adjustable biodegradability, satisfactory flexibility, tensile strength, extension and tear strength, transparency, cloud preventive property, durability, heat and water resistance by taking advantage of specified biodegradable resins and combinations thereof.

Means for Solving the Problems

The present invention provides a biodegradable resin composition comprising at least one stabilizing additive selected from the groups of antioxidants (a), of radical scavenging light stabilizers (b), UV and visible light absorbing compounds (c), and quenchers of photochemical excited states (d). The biodegradable resin composition stabilized with at least one of the aforementioned stabilizing compounds provides a set of materials that can be easily processed to manifold mono- and multilayer film and sheet applications while providing a markable weatherability as well as an enhancement of durability in outdoor applications. For adopting the material to specific applications, e.g. transparent greenhouse films, additional additives like anti-clouding agents (e) and anti-fogging agents (f) may be used as well as other additives improving processability and handling properties of the films and sheets.

The biodegradable resin comprises at least one biodegradable polymer selected from the group consisting of aliphatic (co)polyesters, aromatic-aliphatic (co)polyesters, aliphatic polycarbonates, aromatic-aliphatic polycarbonates, aliphatic polyester-urethanes, partly aromatic polyester-urethanes, aliphatic polyesteramides, aliphatic-aromatic polyesteramides, polyetheresteramides, polysaccharide esters, polysaccharide ether esters such as cellulose or starch derivatives or copolymers and/or mixtures thereof.

The following polymers are preferably suitable:

Aliphatic or partly aromatic polyesters from

A) aliphatic bifunctional alcohols, preferably linear C_2 to C_{10}-dialcohols, such as, for example, ethanediol, butanediol or hexanediol, particularly preferably butanediol, and/or optionally cycloaliphatic bifunctional alcohols, preferably having 5 or 6 C atoms in the cycloaliphatic ring, such as, for example, cyclohexanedimethanol, and/or instead of some or all of the diols, monomeric or oligomeric polyols based on ethylene glycol, propylene glycol or tetrahydrofuran or copolymers thereof having molecular weights of up to 8,000, preferably up to 4,000, and/or optionally small amounts of branched bifunctional alcohols, preferably C_3-C_{12}-alkyldiols, such as, for example, neopentylglycol, and additionally optionally small amounts of alcohols of higher functionality, such as, for example, 1,2,3-propanetriol or trimethylolpropane, and from aliphatic bifunctional acids, preferably C_2-C_{12}-alkyldicarboxylic acids, such as, for example and preferably, succinic acid or adipic acid, and/or optionally aromatic bifunctional acids, such as, for example, terephthalic acid, isophthalic acid or naphthalenedicarboxylic acid, and additionally optionally small amounts of higher functionality, such as, for example, trimellitic acid, or

B) building blocks with acid and alcohol functional groups, preferably having 2 to 12 C atoms in the alkyl chain, for example hydroxybutyric acid, hydroxyvaleric acid or lactic acid, or derivatives thereof, for example å caprolactone or dilactide, or a mixture and/or copolymer of A and B, the aromatic acids making up a content of nor more than 50 wt.%, based on all the acids;

Aliphatic or partly aromatic polyester-urethanes from

C) aliphatic bifunctional alcohols, preferably linear C_2 to C_{10}-dialcohols, such as, for example, ethanediol, butanediol or hexanediol, particularly preferably bunaediol, and/or optionally cycloaliphatic bifunctional alcohols, preferably with a C_5-or C_6-cycloaliphatic ring, such as, for example, cyclohexanedimethanol, and/or, instead of some or all of the diols, monomeric or oligomeric polyols based on ethylene glycol, propylene glycol or tetrahydrofuran or copolymers thereof having molecular weights of up to 4,000, preferably up to 1,000, and/or optionally small amounts of branched bifunctional alcohols, preferably C_3-C_{12}-alkyldiols, such as, for example, neopentylglycol, and additionally optionally small amounts of alcohols of higher functionality, preferably C_3-C_{12}-alkylpolyols, such as, for example, 1,2,3-propanetriol or trimethylolpropane, and from aliphatic bifunctional acids, preferably C_2-C_{12}-alkyldicarboxylic acids, such as, for example and preferably, succinic acid or adipic acid, and/or optionally aromatic bifunctional acids, such as, for example, terephthalic acid, isophthalic acid or haphthalenedicarboxylic acid, and additionally optionally small amounts of acids of higher functionality, such as, for example, trimellitic acid, or

D) building blocks with acid and alcohol functional groups, preferably having 2 to 12 C atoms, for example hydroxybutyric acid, hydroxyvaleric acid or lactic acid, or derivatives thereof, for example å-caprolactone or dilactide, or a mixture and/or a copolymer of C and D,

the aromatic acids making up a content of not more than 50 wt.%, based on all the acids,

E) the reaction product of C and/or D with aliphatic and/or cycloaliphatic bifunctional isocyanates and additionally optionally isocyanates of higher functionality, having preferably 1 to 12 C atoms, or 5 to 8 C atoms in the case of cycloaliphatic isocyanates, e.g. tetramethylene diisocyanate, hexamethylene diisocyanate or isophorone diisocyanate, optionally additionally with linear and/or branched and/or cycloaliphatic bifunctional alcohols and/or alcohols of higher functionality, preferably C_3-C_{12}-alkyldi or –polyols, or 5 to 8 C atoms in the case of cycloaliphatic alcohols, e.g. ethanediol, hexanediol, butanediol or cyclohexanedimethanol, and/or optionally additionally with linear and/or branched and/or cycloaliphatic bifunctional amines and/or amino alcohols and/or amines and/or amino alcohols of higher functionality, having preferably 2 to 12 C atoms in the alkyl chain, e.g. ethylenediamine or aminoethanol, and/or optionally further modified amines of alcohols, such as, for example, ethylenediaminoethanesulfonic acid, as the free acid or as a salt,

the ester content C) and/or D) being at least 75 wt.%, based on the sum of C), D) and E).

Aliphatic or aliphatic polyester-carbonates from

F) aliphatic bifunctional alcohols, preferably linear C_2 to C_{10}-dialcohols, such as, for example, ethanediol, butanediol or hexanediol, particularly preferably butanediol, and/or optionally cycloaliphatic bifunctional alcohols, preferably having 5 to 8 C atoms in the cycloaliphatic ring, such as, for example, cyclohexanedimethanol, and/or instead of some or all of the diols, monomeric or oligomeric polyols based on ethylene glycol, propylene glycol or tetrahydrofuran or copolymers thereof having molecular weights of up to 4,000, preferably up to 1,000, and/or optionally small amounts of branched bifunctional

alcohols, preferably with C_2-C_{12}-alkyldicarboxylic acids, such as, for example, neopentylglycol, and additionally optionally small amounts of alcohols of higher functionality, such as, for example, 1,2,3-propanetriol or trimethylolpropane, and from aliphatic bifunctional acids, such as, for example and preferably, succinic acid or adipic acid, and/or optionally aromatic bifunctional acids, such as, for example, terephthalic acid, isophthalic acid or naphthalenecarboxylic acid, and additionally optionally small amounts of acids of higher functionality, such as, for example, trimellitic acid, or

G) building blocks with acid and alsohol functional groups, preferably having 2 to 12 C atoms in the alkyl chain, for example hydroxybutyric acid, hydroxyvaleric acid or lactic acid, or derivatives thereof, for example å-caprolactone or dilactide, or a mixture and/or a copolymer of F) and G),

the aromatic acids making up a content of not more than 50 wt.%, based on all the acids;

H) a carbonate content which is prepared from aromatic bifunctional phenols, preferably bisphenol A, and carbonate donors, for example phosgene, or a carbonate content which is prepared from aliphatic carbonic acid esters or derivatives thereof, such as, for example, chlorocarbonic acid esters, or aliphatic carboxylic acids or derivatives thereof, such as, for example, salts and carbonate donors, for example phosgene,

the ester content F) and/or G) being at least 70 wt.%, based on the sum of F), G) and H);

Aliphatic or partly aromatic polyester-amides or polyetherester-amides from

I) aliphatic bifunctional alcohols, preferably linear C_2-C_{10}-dialcohols, such as, for example, ethanediol, butanediol or hexanediol, particularly preferably

butanediol, and/or optionally cycloaliphatic bifunctional alcohols, preferably having 5 to 8 C atoms, such as, for example, cyclohexanedimethanol, and/or, instead of some or all of the diols, monomeric or oligomeric polyols based on ethylene glycol, propylene glycol or tetrahydrofuran or copolymers thereof having molecular weights of up to 10,000, preferably up to 8,000, particularly preferably up to 5,000, and/or optionally small amounts of branched bifunctional alcohols, preferably C_3-C_{12}-alkyldiols, such as, for example, neopentylglycol, and additionally optionally small amounts of alcohols of higher functionality, preferably C_3-C_{12}-alkylpolyols, such as, for example, 1,2,3-propanetriol or trimethylolpropane, and from aliphatic bifunctional acids, preferably having 2 to 12 C atoms in the alkyl chain, such as, for example and preferably, succinic acid or adipic acid, and/or optionally aromatic bifunctional acids, such as, for example, terephthalic acid, isophthalic acid or naphthalenedicarboxylic acid, and additionally optionally small amounts of acids of higher functionality, such as for example, trimellitic acid, or

K) building blocks with acid and alcohol functional groups, preferably having 2 to 12 C atoms in the carbon chain, for example hydroxybutyric acid, hydroxyvaleric acid or lactic acid, or derivatives thereof, for example å-caprolactone or dilactide,

or a mixture and/or copolymer of I) and K), the aromatic acids making up a content of not more than 50 wt.%, based on all the acids,

L) an amide content from aliphatic and/or cycloaliphatic bifunctional and/or optionally small amounts of branched bifunctional amines, preferred compounds being linear aliphatic C_2- to C_{10}-diamines, and additionally optionally small amounts of amines of higher functionality, and among the amines preferably hexamethylenediamine or isophoronediamine, and particularly preferably hexamethylenediamine, and from linear and/or cycloaliphatic bifunctional acids, preferably having 2 to 12 C atoms in the alkyl chain, or C_5 or C_6-ring

in the case of cycloaliphatic acids, preferably adipic acid, and/or optionally small amounts of branched bifunctional and/or optionally aromatic bifunctional acids, such as, for example, terephthalic acid, isophthalic acid or naphthalenedicarboxylic acid or naphthalenedicarboxylic acid, and additionally optionally small amounts of acids of higher functionality, preferably having 2 to 10 C atoms, or

M) an amide content of building blocks with acid and amine functional groups, preferably having 4 to 20 C atoms in the cycloaliphatic chain, preferably ù-lauryllactam or å-caprolactam, particularly preferably å-caprolactam, particularly preferably å-caprolactam,

or a mixture of L) and M) as the amide content, the ester content I) and/or K) being at least 30 wt.%, based on the sum ofI), K), L) and M), and preferably the weight content of the ester structures is 30 to 70 wt.% and the content of the amide structures is 70 to 30 wt.%.

Preferred embodiments are aliphatic (co)polyesters, aromatic-aliphatic (co)polyesters, polyetheresteramides and/or aliphatic polyesteramides with the aliphatic polyesteramides being most preferable.

The polyesteramide resin to be used in the present invention has a mean molecular weight of 10,000 to 300,000, preferably 20,000 to 150,000.

The antioxidant (a) is at least one selected from the group of hindered phenol based compounds (a1) and/or phosphite and/or phosphonite ester based compounds (a2) and/or a sulfur containing synergists in antioxidation (a3). The light stabilizer (b) is at least one selected from the hindered amine based light stabilizers, preferably a high molecular weight oligomer or polymer compound comprising at least two repeating units. The UV-absorber (c) is at least one selected from the group of benzophenones, benzotriazoles, benzylidene malonates, oxanilides, benzoaxazinones or

triazines, preferred benzophenones, benzotriazoles. The quencher (d) is selected from the metal organic compounds.

The invention relates to a resin composition wherein

0 – 1,0 preferred 0,1 to 0,8 parts by weight of c),

0 – 1,2, preferred 0,01-0,5 parts by weight of a) 0-1,0, preferred 0,1-0,8 parts by weight of

b) and polymer add up to 100 parts by weight, optionally d) and f) may be added.

The above selection is not limited to compounds providing only one stabilizing mechanism but includes compounds comprising chemically different functional groups each acting as stabilizers but following different mechanisms. Combinations of different stabilizers being selected from only one of the groups (a) to (d) may be applied as well as combinations across those groups.

Examples of the hindered phenol based compounds (a1) to be used in the present invention are octadecyl-3-(3,5-di-t-butyl-4-hydroxyphenyl)propionate, pentadecyl-3-(3-t-butyl-5-methyl-4-hydroxyphenyl)propionate, triethyleneglycol-bis[3-(3-t-butyl-5-methyl-4-hydroxyphenyl)propionate], 1,6-hexanediol-bis[3-(3,5-di-t-butyl-4-hydroxyphenyl)propionate], pentaerythritol-tetrakis[3-(3,5-di-t-butyl-4-hydroxyphenyl)propionate], 1,3,5-trimethyl-2,4,6-tris(3,5-di-t-butyl-4-hydroxybenzyl)-benzene, N,N-hexamethylenebis(3,5-di-t-butyl-4-hydroxy-hydrocinnamide), 2,2-thio-diethylenebis[3-(3,5-di-t-butyl-4-hydroxyphenyl)propionate], 6,6'-di-tert-butyl-2,2'-methylenedi-p-cresol, 6,6'-di-tert-butyl-4,4'-diethyl-2,2'-methylenediphenol, 3,5-di-t-butyl-4-hydroxy-benzylphosphonate diethylester, tris(3,5-di-t-butyl-4-hydroxy-benzyl)isocyanurate, 3,9-bis(1,1-dimethyl-2-[β-(3-t- butyl-4-hydroxy-5-methylphenyl)propionyloxy]ethyl)-2,4,8,10- tetraoxaspiro-(5,5)undecane, tetrakis[methylene-3-(3',5'-di-t-butyl-4'-hydroxyphenyl)propyl]methane, 1,1,3-tris(2-methyl-4-hydroxy-5-t-butylphenyl)butane, and 4,4'-butylidenebis(3-methyl-6-t-butylphenyl).

Examples of the phosphite and/or phosphonite ester based compounds (a2) to be used in the present invention are 2,2-methylenebis[(4,6-di-t-butylphenyl)octylphosphite, tris(2,4-di-t-butylphenyl)phosphite, 1,1,3-tris(2-methyl-4-ditridecylphosphite-5-t-butylphenyl)butane, dinonylphenyl pentaerythritol diphosphite, distearyl pentaerythritol diphosphite, bis(2,4-di-t-butylphenyl)pentaerythritol di-phosphite, bis(2,6-di-t-butyl-4-methylphenyl) phosphite, pentaerythritol di-phosphite, diphenyldecyl phosphite, triphenyl phosphite, tris-nonylphenyl phosphite, tridecyl phosphite, tris(2-ethylhexyl)phosphite, tributyl phosphite, tris(dinonylphenyl)phosphite, trilauryl trithiophosphite, trilauryl phosphite, bis(neopentylglycol)-1,4-cyclohexane dimethylphosphite, distearyl pentaerythritol diphosphite, diisodecyl pentaerythritol diphosphite, tris(lauryl-2-thioethyl)phosphite, tetratridecyl-1,1,3-tris(2'-methyl-5'-t-butyl-4'-oxyphenyl)butane diphosphite, tris(4-oxy-2,5-di-t-butylphenyl)phosphite, tris(4-oxy-3,5-di-t-butylphenyl)phosphite, 2-ethylhexyl diphenylphosphite, tris (mixed mono- and di-nonylphenyl)phosphite, hydrogenated 4,4'-isopropylidene diphenylpolyphosphite, diphenyl-bis[4,4'-n-butylidenebis(2-t-butyl-5-methylphenol)]thiodiethanol diphosphite, 4,4'-butylidenebis(3-methyl-6-t-butylphenol-di-tridecyl)phosphite, bis(octylphenyl)-bis[4,4'-n-butylidenebis(2-t-butyl-5-methylphenol)]-1,6-hexanediol diphosphite, phenyl-4,4'-isopropylidenediphenol-pentaerythritol diphosphite, phenyldiisodecyl phosphite, tetratridecyl-4,4'-n-butylidenebis(2-t-butyl-5-methyphenol)diphosphite, tris(2,4-di-t-butylphenyl)phosphite, tristearyl phosphite, octyldiphenyl phosphite, diphenyltridecyl phosphite, phenyl-di(tridecyl)phosphite, tris(2-cyclohexylphenyl)phosphite, ditridecyl-di(2-cyclohexylphenyl) hydrogenated bisphenol A diphosphite, di(2,4-di-t-butylphenyl)cyclohexyl phosphite, 2,4-di-t-butylphenyl-diisodecyl phosphite, tris(butoxyethoxyethyl)phosphite, diphenyl acidophosphite, bis(2-cyclohexylphenyl)acidophosphite, bis(2,4-di-t-butylphenyl)acidophosphite, bis(nonylphenyl)acidophosphite, dibenzyl acidophosphite, and tetrakis(2,4-di-t-butylphenyl)- 4,4'-biphenylene phosphite.

Examples of the sulfur containing compounds (a3) are didodecyl 3,3'-thiodipropionate, di(tridecyl) 3,3'-thiodipropionate, and dioctadecyl 3,3'-thiodipropionate.

Examples of the radical scavenging light stabilizer (b) to be used in the present invention are poly[(6-morpholino-s-triazine-2,4-diyl)[2,2,6,6-tetramethyl-4-piperidyl)imino]-hexamethylene[(2,2,6,6-tetramethyl-4-piperidyl)imino]], the polymer of N,N'-bis(2,2,6,6-tetramethyl-4-piperidinyl-1,6-hexanediamine with morpholine-2,4,6-trichloro-1,3,5-triazine, poly ⟨[6-(1,1,3,3-tetramethylbutyl)amino-1,3,5-triazin-2,4-diyl]-[(2,2,6,6-tetramethyl-piperidyl)imino]hexamethylene [(2,2,6,6-tetramethyl-piperidyl)imino]⟩, poly ⟨[6-(1,1,3-trimethylpentyl)imino-1,3,5-triazine-2-4-diyl]-[(N-methyl-2,2,6,6-tetramethyl-piperidyl)imino]octamethylene-[(N-methyl-2,2,6,6-tetramethyl-piperidyl)imino]⟩, 2,2,6,6-tetramethylpiperidinyl-4-benzoate, bis-(2,2,6,6-tetramethyl-4-piperidinyl)sebacate, 1,3,8-triaza-7,7,9,9-tetramethyl-3-n-octyl-spiro[4,5]decane-2,4-dione, 1,2,3,4-tetra(4-carbonyloxy-2,2,6,6-tetramethyl-piperidine)-butane, tri-(4-acetoxy-2,2,6,6-tetramethyl-piperidine)-amine, 4-acetoxy-2,2,6,6-tetramethylpiperidine, 4-stearoylpxy-2,2,6,6-tetramethylpiperidine, 4-benzyloxy-2,2,6,6-tetramethylpiperidine, 4-(phenylcarbamoyloxy)-2,2,6,6- tetramethylpiperidine, tris(2,2,6,6-tetramethyl-4-piperidine) phosphite, bis(2,2,6,6-tetramethyl-4-piperidyl)terephthalate, 1,3,8-triaza-7,7,9,9-tetramethyl-2,4-dioxo-spiro-[4,5]decane, (2,2,6,6-tetramethylpiperidine)-4-spiro-2'-(6', 6'-dimethylpiperidine)-4'-spiro-5"-hydantoin, bis(1,2,2,6,6-pentamethyl-4-piperidyl)sebacate, 3-dodecyl-1-(2,2,6,6-tetramethyl-4-piperidyl)pyrrolidine-2,5-dione, polmethyl8propyl-3-oxy(2,2,6,6-tetramethyl-4-piperidinyl)]siloxane, the polycondensation product of dimethyl succinate with 4-hydroxy-2,2,6,6-tetramethyl-1-piperidine ethanol, tetrakis(2,2,6,6-tetramethyl-4-pyperidyl)-1,2,3,4-butane tetracarboxylate, and tetrakis(1,2,2,6,6-pentamethyl-4-piperidyl)-1,2,3,4-butane tetracarboxylate.

Examples of the UV-absorbers (c) being used in the present invention are 2-hydroxy-4-octyloxy-benzophenone, 2-hydroxy-4-methoxy-benzophenone, the reaction product of methyl 3-(3-(2H-benzotriazol-2-yl)-5-t-butyl-4-hydroxyphenyl)-propionate with PEG 300, 2-(2H-benzotriazol-2-yl)-4,6-bis(1-methyl-1-phenylethyl)phenol, 2-(2H-benzotriazol-2-yl)-4,6-di-t-butylphenol, 2-(5-chloro-2H-benzotriazol-2-yl)-6-t-butyl-4-methyl-phenol, 2,4-di-t-butyl-6-(5-chloro-2H-benzotriazol-2-yl)-phenol, 2-(2H-benzotriazol-2-yl)-4,6-di-t-pentylphenol, 2-(2H-benzotriazol-2-yl)-4-(1,1,3,3-

tetramethylbutyl)phenol, 2-(2H-benzotriazol-2-yl)-4-t-butyl-6-(1-methylpropyl)-phenol, 2,2'-methylene-bis(6-(2H-benzotriazol-2-yl)-4-(1,1,3,3-tetramethylbutyl)-phenol), 2-(2H-benzotriazol-2-yl)-6-dodecyl-4-methylphenyl where the dodecyl moiety is branched or linear, 2-(4,6-diphenyl-1,3,5-triazin-2-yl)-5-hexyloxy-phenol, 2-(2H-benzotriazol-2-yl)-p-cresol, 2-[4,6-bis(2,4-dimethylphenyl)-1,3,5-triazin-2-yl]-5-octyloxy-phenol, 2,2'-(1,4-phenylene)bis-4H-3,1-Benzoxazin-4-one, N-(2-ethoxy-phenyl)-N'-(2-ethylphenyl)-ethanediamide, N-(2-ethoxyphenyl)-N'-(4-isodecyl-phenyl)-ethanediamide, and dimethyl 4-methoxybenzylidene malonate.

An examples of a quencher (d) being used in the present invention is [2,2'-thio-bis(4-t-octylphenolato)]-n-butylamine nickel(II).

The anti-clouding agent (e) being used in the present invention is selected from the groups of partial esters of a polyfunctional alcohol or from the group of the condensation products of the polyfunctional alcohol with a higher fatty acid. The cloud preventive agents include, for example, a sorbitane based surfactant such as sorbitan monostearate and sorbitan monopalmitate, a glycerine based surfactant such as glycerine monopalmitate and diglycerine monopalmitate, and a polyethylene glycol based surfactant such as polyethyleneglycol monostearate, polyethylene glycol monopalmitate and polyethylene glycol alkylphenyl ether, a trimethylol propane based surfactant and a pentaerythritol based surfactant, and an isomer or an alkylene oxide addition compound thereof.

Examples of the anti-fogging agent (f) being used in the present invention and preventing fog from generating in the vicinity of house cover films are fluorinated surfactants or silicone based surfactants.

Description of the Embodiments

The present invention provides a biodegradable film, preferred agricultural film in which at least one of the additives (a) to (f) are blended into a resin composition

mainly composed of a biodegradable resin which is preferably an aliphatic polyester and/or copolyester and/or an aromatic-aliphatic copolyester and/or an aliphatic polyesteramide with the aliphatic polyesteramide being the most preferred one.

The biodegradable resin composition is prepared by blending relative to 100 parts by weight of the composition 0.03 to 1.2 parts by weight, preferably in the range of 0.05 to 1.0 parts by weight of an antioxidant (a), more preferably a hindered phenol based compound (a1) and/or a phosphite and/or phosphonite ester (a2). It is not preferable that these ratios are smaller than the range described above, since heat stability tends to be decreased. It is also not preferable that the ratios are larger than the range described above, since appearance of the molded product is liable to be poor, or the product cost may be increased.

The biodegradable resin composition may also comprise a hindered amine based light stabilizer (b) in an amount of 0.03 to 1.0 parts by weight, preferably in the range of 0.05 to 0.8 parts by weight, relative to 100 parts by weight of the composition. The blending amounts of lower and higher than the ranges described above are not preferable, since weather resistance is decreased in the former case, and appearance of the product becomes poor or the production cost tend to increase in the latter case.

The biodegradable resin composition may also comprise an UV-absorber (c) preferably a benzophenone and/or a benzotriazole in an amount of 0.01 to 1.0 parts by weight, preferably in the range of 0.05 to 0.5 parts by weight, relative to 100 parts by weight of the composition.

Other biodegradable polymer materials may be added to the formulation as described above in a range not inhibiting the effect of the present invention. It is also possible to add additives and reforming agents such as a sulfur based antioxidant (a3), a Ni based quencher (d), an anti-clouding agent (e), an anti-fogging agent (f), a plasticizer, filler, lubricant, reinforcing agent, inorganic heat insulator, antifungal agent, pigment

and fluorescent substance in order to control forming ability and other properties of the film and sheet.

When the composition according to the present invention is formed into a film or sheet for general purpose, preferably consisting of a monolayer, the thickness is usually in the range of 15 to 1000 µm, preferably in the range of 20 to 500 µm.

In the present invention for agricultural films and sheets of improved weatherabilty that are preferably transparent the preferred composition of the biodegradable resin comprises a blend of a biodegradable polyesteramide, preferably an aliphatic polyesteramide, and a biodegradable polyester, preferably an aliphatic polyester. It was found that the polyester present in these blends provided a diffusion barrier for water migrating into the polyesteramide phase. Thus photochemical initiation and photochemical formation of reactive species being responsible for chain scission reactions where water is actively involved in the reaction mechanism are markably retarded. The preferred resin composition being used for transparent agricultural films and sheets comprises 60 to 95 parts by weight of the polyesteramide and 5 to 40 parts by weight of the aliphatic polyester.

The amount of the hindered amine based light stabilizer being added to this composition is in the range of 0.03 to 1.0 parts by weight, preferably in the range of 0.05 to 0.5 parts by weight.

It is not preferable that the blending amount is smaller than the range described above, since weather resistance tends to decrease while, when the amount is larger, appearance of the molded product is compromised or the production cost may be increased.

Other biodegradable polymer materials may be added to the formulation as described above in a range not inhibiting the effect of the present invention. It is also possible to add additives and reforming agents such as antioxidants (a), an UV-absorber (c), a

Ni based quencher (d), an anti-clouding agent (e), an anti-fogging agent (f), a plasticizer, filler, lubricant, reinforcing agent, inorganic heat insulator, antifungal agent, pigment and fluorescent substance in order to control forming ability and other properties of the film and sheet.

The film thickness is in the range of 20 to 200 μm, preferably in the range of 40 to 150 μm.

For multilayer films and sheets it is preferable in the present invention to use a resin composition comprising 60 to 100 parts by weight, preferably 80 to 100 parts by weight of the polyesteramide resin and 0 to 40 parts by weight, preferably 0 to 20 parts by weight of the aliphatic polyester resin.

The ratio of the hindered phenol based antioxidant (a1) and/or phosphite and/or phosphonite ester based antioxidant (a2) is in the range of 0.01 to 1.0 parts by weight, preferably in the range of 0.03 to 0.5 parts by weight, relative to 100 parts by weight of the resin composition.

An amount of larger than 0.01 parts by weight of an antioxidant improves durability. However, a larger amount than 1.0 parts by weight is not preferable since appearance of the molded product is compromised or the production cost may be increased.

Hindered amine based light stabilizers (b) are added into the composition for manufacturing agricultural multilayer films of the biodegradable resin, if necessary.

The ratio of the hindered amine based light stabilizer (b) is in the range of 0.01 to 1.0 parts by weight, preferably in the range of 0.05 to 0.5 parts by weight relative to 100 parts by weight of the resin composition.

Examples of the inorganic fillers and/or colorants to be used in the multilayer films include hydrophobic inorganic fillers such as silica, talc, clay, calcium carbonate,

magnesium carbonate, titanium oxide and alumina; and inorganic and organic colorlants such as carbon black. The latter one being used for non-transparent mulching films only. These additives are blended in a range of 0.5 to 50 parts by weight, preferably in the range of 1 to 40 parts by weight, relative to 100 parts by weight of the resin composition.

Other biodegradable polymer materials may be added to the formulation as described above in a range not inhibiting the effect of the present invention. It is also possible to add additives and reforming agents such as an UV-absorber (c), a Ni based quencher (d), an anti-clouding agent (e), an anti-fogging agent (f), a plasticizer, filler, lubricant, reinforcing agent, inorganic heat insulator, antifungal agent, pigment and fluorescent substance in order to control forming ability and other properties of the film and sheet.

The film thickness is in the range of 15 to 100 μm, preferably in the range of 20 to 50 μm in order to provide fast biodegradation.

For utilizing the biodegradable resin compositions described above in specific applications and meeting all requirements of agricultural polytunnel or greenhouse applications, multilayer films comprising combinations of materials with each layer having its specific function are developed.

The number of layers in the biodegradable multilayer film according to the present invention is not especially limited. The film is composed of at least two layers in the order of, for example, (A)/(B), (B)/(A)/(B), (B)/(A')/(A)/(B) or (B)/(A')/(A)/(A')/(B). The layer (A) has characteristics such as transparency, flexibility and expansion, while the layer (B) contributes to improvement of water resistance. Therefore, it is preferable that both outer layers of the biodegradable multilayer film according to the present invention are composed of the layers (B), and the layers (A) and (A') constitute the intermediate layers, in order to render characteristics of the biodegradable multilayer film to be excellent in transparency, flexibility and water resistance.

In a preferred embodiment of the present invention the substrate layer (A) mainly composed of a polyesteramide resin, preferably an aliphatic polyester resin, is prepared by blending the polyesteramide resin in a range of 50 to 100% by weight, preferably in a range of 60 to 95% by weight, and a biodegradable polyester resin, preferably an aliphatic polyester resin, in a range of 0 to 50% by weight, preferably in a range of 5 to 45% by weight. A blending amount of the polyesteramide resin of less than 50% by weight is not preferable since transparency, flexibility and extension tend to decrease.

A surface layer (B) mainly composed of an polyester resin, preferably an aliphatic polyester resin, is provided on at least one face of the substrate layer (A). The blending ratios of the aliphatic polyester resin in the surface layer (B) are in the range of 60 to 100% by weight, preferably in the range of 75 to 100% by weight, and that of the polyesteramide resin is in the range of 0 to 40% by weight, preferably in the range of 0 to 25% by weight, respectively. It is not preferable that the blending ratio of the aliphatic polyester resin is less than 60% by weight since water resistance is deteriorated.

The thickness of the layer (A) is 50 to 90%, preferably 60 to 85%, of the overall thickness of the film. When the thickness of the layer (A) is less than 50%, flexibility and transparency becomes poor while, when the thickness exceeds 90%, water resistance of the layer tends to be poor due to reduced thickness of the layer (B).

Other biodegradable polymer materials may be added to the formulation as described above in a range not inhibiting the effect of the present invention. It is also possible to add additives and reforming agents such as antioxidants (a), UV-stabilizers (b), UV-absorbers (c), a Ni based quencher (d), an anti-clouding agent (e), an anti-fogging agent (f), a plasticizer, filler, lubricant, reinforcing agent, inorganic heat insulator, antifungal agent, pigment and fluorescent substance in order to control forming ability and other properties of the film and sheet.

The thickness of the multilayer film, formed by using the compositions for respective layers according to the present invention, is usually in the range of 20 to 2000 µm, preferably in the range of 40 to 500 µm.

In order to meet the technical standards of greenhouse and polytunnel applications with respect to anti-clouding and anti-fogging property the biodegradable multilayer agricultural films according to the present invention may be composed of at least three layers. For example, the layer construction may be such as (A)/(B)/(C), (A)/(C)/(B')/(C) and (A)/(B)/(B')/(B")/(C). The layer (A) has characteristics such as water resistance and dust preventive property, while the layers (B), (B') and (B") contribute for improving transparency, duration of cloud preventive property, and flexibility. The layer (C) serves for maintaining and duration of some cloud preventive agent. Accordingly, the completely biodegradable multilayer agricultural film according to the present invention is excellent in the characteristics such as transparency, flexibility and cloud preventive property.

In the layer (A) mainly composed of a polyester, preferably an aliphatic polyester, the blending ratio of the polyester resin is in the range of 60 to 100% by weight, preferably in the range of 75 to 95% by weight, and the blending ration of a polyesteramide resin, preferably an aliphatic polyesteramide resin, is 0 to 40% by weight, preferably 5 to 25% by weight. It is not preferable that the blending ratio of the polyester resin is less than 60% by weight, since water resistance is decreased.

In the layer (B) mainly composed of a polyesteramide resin, preferably an aliphatic polyesteramide resin, the blending ratio of the polyesteramide resin is in the range of 50 to 100% by weight, preferably in the range of 60 to 95% by weight, and the blending ratio of the polyester resin is in the range of 0 to 50% by weight, preferably in the range of 5 to 40% by weight. One to four parts by weight of a cloud preventive agent (e) is blended relative to 100 parts by weight of the resin composition. It is not preferable that the blending ratio of the polyesteramide resin is less than 50% by

weight, since duration of cloud preventive property, transparency, flexibility and extension tend to be decreased.

In the layer (C) mainly composed of a polyester, preferably an aliphatic polyester resin, the blending ratio of the polyester resin is in the range of 60 to 95% by weight, preferably in the range of 70 to 90% by weight, and the blending ratio of the polyesteramide resin, preferably an aliphatic polyesteramide resin, is in the range of 5 to 40% by weight, preferably in the range of 10 to 30% by weight according to the present invention. Into 100 parts by weight of the resin composition, 0.1 to 2 parts by weight of the cloud preventive agent (e) is blended. It is not preferable that the blending ratio of the polyester resin is less than 60% by weight, since water resistance becomes poor.

At least 5% by weight of the sub-component resins are blended to the principal component resin in at least one of the layer (A) or layer (B). It is not preferable that the blending ratio is smaller than the ratio described above, since adhesive property becomes poor. The thickness of the layer (B) is 45 to 90%, preferably 55 to 80%, of the overall thickness of the film. The thickness of the layer (B) of less than 45% results in poor flexibility, transparency and duration of cloud preventive property. It is not preferable, on the other hand, that the thickness exceeds 90%, since water resistance of the biodegradable agricultural film tends to deteriorate because the thicknesses of the water resistant layers (A) and (C) are reduced.

The preferable cloud preventive agent (e) to be blended in the layers (B) and (C) of the biodegradable multilayer agricultural film according to the present invention is a partial ester of a polyfunctional alcohol or a condensation product of the polyfunctional alcohol with a higher fatty acid.

A so-called fog preventive property may be provided by adding an anti-fogging agent (f) such as fluorinated surfactants and silicone based surfactant for preventing fog generating in the vicinity of the inner face of the house cover.

For maintaining good weatherability stabilizers such as a hindered phenol based compound (a1) and/or phosphite and/or phosphonite ester based compound (a2) are blended in the range of 0.03 to 1.2 parts by weight, preferably in the range of 0.05 to 0.8 parts by weight, relative to 100 parts by weight of the resin components in each layer. When the amount of stabilizers blended into the composition is smaller than the proportion described above, heat stability tends to be decreased while, when the amount is larger, appearance of the molded products is compromised or results in cost-up of the product.

Also a hindered amine based light stabilizer (b) is blended in the range of 0.03 parts by weight, preferably in the range of 0.05 to 0.5 parts by weight, relative to 100 parts by weight of the resin components in each layer. It is not preferable that the amount of blending is smaller than the proportion described above since weather resistance is deteriorated while, when the proportion is larger, appearance of the molded products is compromised or the production cost is increased.

Other biodegradable polymer materials may be added to the formulation as described above in a range not inhibiting the effect of the present invention. It is also possible to add additives and reforming agents such as sulfur containing synergists in antioxidantion (a3), UV-absorbers (c), a Ni based quencher (d), a plasticizer, filler, lubricant, reinforcing agent, inorganic heat insulator, antifungal agent, pigment and fluorescent substance in order to control forming ability and other properties of the film and sheet.

The thickness of the multilayer film, formed with the compositions in each layer according to the present invention, is usually in the range of 20 to 200 μm, preferably in the range of 40 to 150 μm.

Also it has been found in the present invention that if inorganic particles are blended to the bi degradable resin composition the biodegradability of the respective films

and sheets can be controlled. The inorganic particles to be used in the present invention preferably include those having hydrophobicity such as silica, talc, clay, calcium carbonate, calcium sulfate, magnesium carbonate, titanium oxide and alumina. Blending the hydrophobic inorganic particles allows permeation of water and hydrolysis of the resin at the initial stage of decomposition to be enhanced, thereby accelerating overall biodegradation reaction. Although the particle size is not limited, a larger specific surface area is preferable for enhancing collapse effects. The blending ratio is 2.5 to 50 parts by weight, preferably 3.5 to 40 parts by weight relative to 100 parts by weight of the resin composition. When the content of the inorganic particles is less than 2.5 parts by weight, degradability is not enhanced while, when the content is larger than 50 parts by weight, physical properties of the molded body, for example tensile strength, is decreased to compromise practical applicability.

The provided film and sheet, and a molded body comprising the film or sheet having biodegradability obtained as described above can be obtained by hot molding methods such as vacuum molding and compressed air molding, which can be used for manufacturing a blister processing body, and a food cup and tray. The film or sheet can be also used for manufacturing a box-type package by ruling for bending the sheet. Further, the resin composition can be also used for a sheet or molded body laminated with a sheet of paper and other plastic films.

When the composition according to the present invention is formed into a film or sheet, the thickness is usually in the range of 15 to 1000 µm, preferably in the range of 20 to 500 µm.

In a preferred embodiment of the present invention the biodegradable multilayer films described above are used in rice fields. Herbicides have been sprayed in rice cultivation for preventing weeds from budding. Since the herbicides may adversely affect human bodies and may be an important factor of environmental pollution, there are some rice fields where agricultural chemicals such as herbicides are not used at all. However, manually removing the weeds cost a person so much labor that

weed removing methods that can save man's labor with no effects on human bodies have been desired.

The present invention further provides a method for planting young rice plants comprising spreading the multi-film of the biodegradable resin for use in rice fields according to the present invention over a rice field, and planting the young rice plant by piercing the multi-film.

It is preferable to use 60 to 100 parts by weight, preferably 80 to 100 parts by weight, of the polyester amide resin and 0 to 40 parts by weight, preferably 0 to 20 parts by weight, of the aliphatic polyester resin.

The resin composition for this application mainly composed of a polyesteramide comprises additives such as a hindered phenol based compound (a1) and/or a phosphite and/or phosphonite ester based compound (a2), and an inorganic filler and/or a colorant.

The hindered phenol based compound (a1) and/or phosphite and/or phosphonite ester based compound (a2) are blended in the range of 0.01 to 1.0 parts by weight, preferably in the range of 0.03 to 0.5 parts by weight, relative to 100 parts by weight of the resin components. When the amount of blending is larger than 0.01 parts by weight, durability can be improved. However, when the amount is too large, appearance of the molded products is compromised or results in cost-up of the product.

A hindered amine based light stabilizer (b) may be blended to the multi-film of the biodegradable resin for use in rice fields, if necessary. The amount of the hindered amine based light stabilizer (b) is in the range of 0.01 to 1.0 part by weight, preferably 0.03 to 0.5 parts by weight, relative to 100 parts by weight of the resin component.

Example of inorganic fillers and/or colorants to be used in the present invention include hydrophobic inorganic fillers such as silica, talc, clay, calcium carbonate, magnesium carbonate, titanium oxide and alumina, and inorganic and organic colorants such as carbon black. The amount of these additives is in the range of 0.5 to 50 parts by weight, preferably in the range of 1 to 40 parts by weight, relative to 100 parts by weight of the resin component.

The thickness of the film in this mulching application is usually in the range of 15 to 100 μm, preferably in the range of 20 to 50 μm.

Methods of preparation

While the biodegradable resin comprising the biodegradable polymers and additives (a) to (f) described above may be mixed during the extrusion process, it is recommended to previously formulate each ingredient before forming pellets. The methods for formulating the resin components and various additives are not especially limited, but any methods that have been used for preparing conventional plastic compositions, for example heat-melting and kneading using a kneading machine such as a kneader, Banbury mixer and roll, a mixing machine such as ribbon blender and Henshel mixer, and uniaxial or biaxial extruder, may be used.

The biodegradable films according to the present invention are manufactured, for example, by forming a film by a conventional extrusion-molding such as a T-die method or inflation method, followed by cooling with a cooling roll, water or air. Use of a calendar molding is also possible.

Co-extrusion method is preferably used in producing the biodegradable multilayer film according to the present invention. Actually, the layers are melt-extruded with plural extruders corresponding to the lamination number to laminate the molten resin by a conventional method such as a T-die method or inflation method, followed by cooling with a cooling roller, water or air.

Biodegradable films and sheets are produced as described from the composition according to the present invention above, and are used for various applications.

Examples

The present invention is not limited in any sense by the examples as set forth hereinafter. Measurements and evaluations in the examples were carried out under the following conditions.

1) Weather resistance

Weather resistance (a):
Samples were placed in Sunshine W.O.H and the time lapse before the samples are broken by being degraded in the environment was measured.

Weather resistance (b):
Samples were placed in Sunshine W.O.H by attaching an aluminum plate on the back face of each sample, and the time lapse before the samples are broken by being degraded in the environment was measured.

2) Weatherometer test
Samples were placed in a weatherometer SUN-WOM (made by Suga Test Machine Co. or by Gas Test Equipment Co.) and the time required for exhibiting weather deterioration of the samples was measured:

O: 300 hours or more than

Δ: 150 hours or more and less than 300 hours

×: less than 150 hours

3) Biodegradability
Preparation of test samples

1. Samples are cut into 6 cm square each and are weighed. The mass per one exposure area (4 cm square) is calculated to estimate the initial weight of the decomposition sample.

2. An aluminum plate with an appropriate thickness is cut into 6 cm square, and a 4 cm square plate is cut out of the central portion of the aluminum plate (an aluminum frame).

3. Double-adhesive tapes are attached to the aluminum frame so as to be able to sandwich the test sample with a pair of the aluminum frames.

4. The mass of tow pair of aluminum frames with double-adhesive tapes are weighed.

5. The test sample sandwiched with the aluminum frame is subjected to a biodegradation test.

Preparation method of a compost to be used in biodegradation test

1. A sealable vessel with an appropriate size is prepared (with a depth of 5 cm or more).

2. A commercially available compost is moisturized before use. Water is added to the compost not to pool in the bottom of the vessel, but so that water slightly seeps out of the compost by holding it with the fingers, which prescribed the standard test condition.

Method of biodegradation test

1. Biodegradation tests are carried out in the compost. The test sample is set at a depth about 3 cm deep from the surface of the prepared compost.

2. After setting the test sample, it is placed in a sealed oven heated at 58°C to start the biodegradation test.

3. The lid of the oven is open at least once a day during the biodegradation test, if possible, for spraying water in order to maintain the initial moisture content in the compost.

4. The sample during biodegradation is occasionally taken out of the oven and, after washing with water and wiping the moisture on the surface of the sample, it is weighed to calculate the biodegradation rate from mass changes.

Evaluation criterion

The days required for 80% or more of weight loss of the sample in the biodegradation test is defined to be the days for biodegradation.

◎: less than 40 days

O: 40 days or more and less than 60 days

Δ: 60 days or more and less than 80 days

×: 80 days or more

4) Haze

Haze values represented by % were measured with a haze meter (made by Suga Test Machine Co.).

5) Cloud preventive property:

The film was extended over a water bath warmed at a temperature of 40°C with a tilt angle of 30 degree at an ambient temperature of 5°C. Cloudiness of the film was measured 30 days after based on the following evaluation criterion:

O: Water drops covered 20% or less of the total film area.

Δ: Water drops covered 20% or more and less than 50% of the total film area.

×: Water drops covered more than 50% of the total film area.

6) Tensile strength:

Tensile strength was measured following JIS K-6732 (in kg/cm^2 unit).

7) Extension:

Extension was measured following JIS K-6732 (%).

8) Perpendicular tear strength

Tear strength was measured following JIS K-6732 (in Kg/cm unit).

9) Water resistance test

The sample is cut out in 10 cm square pieces, and two sheets of these samples were inserted between two aluminum plates, which were fixed at four points with clips to submit as a test sample.

This test sample is dipped in water warmed at 70 °C and, after cooling to room temperature, the sample films are peeled off with each other.

◉: The films are peeled off with no resistance

O: The films are peeled off with little resistance

Δ: The films experience some resistance, but can be peeled off with difficulty.

×: The films are not peeled off at all.

10) Flexibility (tensile elasticity modulus)

The tensile elasticity modulus was measured according to ASTM D-882 (in kg/mm^2 unit).

11) Wrinkles and slacks of the moist film

Wrinkles and slacks of the film extended over the agricultural house are observed when the inside surface of the film is moist.

◎: No wrinkles and slacks

O: Substantially no wrinkles and slacks

Δ: A few wrinkles and slacks

×: Marked wrinkles and slacks

12) Cloud preventive blocking property (performance of ventilation)

The sample with a dimension of 25 cm in length and 5 cm in width is adhered on the back face of the lid, which is placed over a water bath warmed at 40°C for 24 hours so that the sample film comes downward of the lid. Then, the wet sample is wound around a glass tube with a diameter of 1 cm to leave the tube in an oven heated at 70°C for 24 hours. The sample taken out of the oven is further left in a constant temperature chamber at 23°C.

The sample wound around the galss tube is peeled off and its surface resistance is measured with a surface measurement apparatus (made by New TOYO Scientific Instrument Co., trade name HEIDON-14).

O: less than 20 g/5 cm

Δ: 20 g/5 cm or more and less than 80 g/5 cm

×: more than 80 g/5 cm

13) Tensile strength (moist film)

After folding the film in water at 15°C for 1 hour, the tensile strength of the moist film was measured according to JS K-6732 (in kg/cm^2 unit).

14) Extension (moist film)

After folding the film in water at 15°C for 1 hour, extension of the moist film was measured according to JS K-6732 (%).

15) Flexibility (extension elasticity modulus) (moist film)
After folding the film in water at 15°C for 1 hour, the extension elasticity modulus of the moist film was measured according to JS D-882 (in kg/mm^2 unit).

Example 1.1

Pellets were produced by blending 0.2 parts by weight of a hindered phenol based compound (made by Asahi Electrochemical Co., trade name; AO-80) and 0.1 parts by weight of a phosphite ester based compound (made by Asahi Electrochemical Co., trade name; "2112") to 100 parts by wight of a polyesteramide resin (melting point: 137°C), prepared by polymerization of adipic acid, 1,4-butanediol and ε-caprolactam, comprising 50% by weight of an aliphatic ester unit and 50% by weight of an aliphatic amide unit. A film with a thickness of 100 μm was formed from the pellets using a T-die type extruder equipped with a cylinder with a diameter of 30 mm. This film was evaluated as 120 hours under the weather resistance test conditions (a), and 100 hours under the weather resistance test conditions (b).

Comparative Example 1.1

A film was prepared by the same method as used in Example 1.1, except that the hindered phenol based compound and phosphite ester based compound used in Example 1 were not blended.

This film was evaluated as 80 hours under the weather resistance test conditions (a), and 50 hours under the weather resistance test conditions (b).

Example 1.2

Pellets were produced by blending 0.1 parts by weight of the hindered phenol basd compound (made by Asahi Electrochemical Co., trade name: AO-60), 0.1 parts by weight of the phosphite ester based compound (made by Asahi Electrochemical Co., trade name: "2112") and 0.3 parts by weight of the hindered amine based light stabilizer (made by Ciba Speciality Chemical Co., trade name: LS-944) to 100 parts by weight of the polyesteramide resin used in Example 1.1. A film with a thickness of 100 µm was formed from the pellets using a T-die type extruder equipped with a cylinder with a diameter of 30 mm.

This film was evaluated as 250 hours under the weather resistance test conditions (a), and 180 hours under the weather resistance test conditions (b).

Example 1.3

Pellets were produced by blending 0.1 parts by weight of the hindered phenol based compound (made by Asahi Electrochemical Co., trade name AO-80), 0.1 parts by weight of the phosphite ester based compound (made by Asahi Electrochemical Co., trade name: "2112"), 0.3 parts by weight of the hindered amine based light stabilizer (made by Ciba Speciality Chemical Co., trade name: LS-944) and 0.1 parts by weight of a benzotriazole based UV absorbing agent (made by Kyodo Pharmacuticals Co., trade name: VS-550) to 100 parts by weight of the polyesteramide resin used in Example 1.1. A film with a thickness of 100 µm was formed from the pellets using a T-die type extruder equipped with a cylinder with a diameter of 30 mm.

This film was evaluated as 350 hours under the weather resistance test conditions (a), and 190 hours under the weather resistance test conditions (b).

Comparative Example 1.2

A film was prepared by the same method as used in Example 1.3, except that only 0.2 parts by weight of the benzotriazole based UV absorbing agent (made by Kyodo Pharmacuticals Co., trade name: VS-550) used in Example 1.3 was blended.

This film was evaluated as 100 hours under the weather resistance test conditions (a), and 60 hours under the weather resistance test conditions (b).

Example 1.4

A film with a thickness of 100 µm was prepared by the same method as used in Example 1.1, except that the polyesteramide resin (melting point: 116 °C), produced by polymerization of adipic acid, 1,4-butanediol and ε-caprolactam, comprising 70% by weight of the aliphatic amide unit and 30% by weight of the aliphatic fatty acid unit was used.

This film was evaluated as 130 hours under the weather resistance test conditions (a), and 100 hours under the weather resistance test conditions (b).

Example 1.5

Pellets were produced by blending 0.1 parts by weight of the hindered phenol based compound (made by Asahi Electrochemical Co., trade name : AO-60), 0.1 parts by weight of the phosphite ester based compound (made by Asahi Electrochemical Co., trade name "2112") and 0.3 parts by weight of the hindered amine based light stabilizer (made by Ciba Speciality Chemical Co., trade name: LS-944) to 100 parts by weight of the polyesteramide resin (melting point: 172°C), produced by polymerization of adipic acid, 1,4-butanediol and hexamethylene diamine, comprising 80% by weight of the aliphatic ester unit and 20% by weight of the aliphatic amide unit. A

film with a thickness of 100 μm was prepared from the pellets using the T-die type extruder having a cylinder with a diameter of 30 mm.

This film was evaluated as 150 hours under the weather resistance test conditions (a), and 120 hours under the weather resistance test conditions (b).

Comparative Example 1.3

A film was prepared by the same method as used in example 1.5, except that the hindered phenol based compound and phosphite ester based compound used in Example 1.5 were not blended.

This film was evaluated as 90 hours under the weather resistance test conditions (1), and 70 hours under the weather resistance test conditions (2).

Example 2.1

Pellets were formed by blending 10 parts by weight of calcium carbonate (made by Maruo Calcium Co., trade name: Super SS, mean grain size: 2.2 μm) relative to 100 parts by weight of a polyesteramide resin (melting point: 137 °C), produced by polymerization of adipic acid, 1,4-butanediol and ε-caprolactam, comprising 50% by weight of an aliphatic ester unit and 50% by weight of an aliphatic amide unit. A film with a thickness of 30 μm was formed from the pellet using a T-die type extruder equipped with a cylinder with a diameter of 30 mm. The days required for biodegradation of this film were 35 days.

Example 2.2

A film was formed by the same method as used in Example 2.1, except that the amount of blending of calcium carbonate in Example 2.1 was changed to 40 parts by weight. The days required for biodegradation of this film were 25 days.

Example 2.3

A film was formed by the same method as used in Example 2.1, except that the amount of blending of calcium carbonate in Example 2.1 was changed to 70 parts by weight. The days required for biodegradation of this film were 20 days.

Example 2.4

A film was formed by the same method as used in Example 2.2, except that 40 parts by weight of talc (made by Fuji Talc Co., mean grain size: 1.8 μm) was used in place of calcium carbonate used in Example 2.2. The days required for biodegradation of this film were 27 days.

Comparative Example 2.1

A film was formed by the same method as used in Example 2.1, except that calcium carbonate used in Example 2.1 was not blended at all. The days required for biodegradation of this film were 45 days.

Example 2.5

Pellets were formed by blending 40 parts by weight of calcium carbonate (made by Maruo Calcium Co., trade name: Super SS, mean grain size: 2.2 μm) relative to 100 parts by weight of a polyesteramide resin (melting point: 116°C), produced by polymerization of adipic acid, 1,4-butanediol and ε-caprolactam, comprising 70% by weight of an aliphatic ester unit and 30% by weight of an aliphatic amide unit. A film with a thickness of 30 μm was formed from the pellet using a T-die type extruder equipped with a cylinder with a diameter of 30 mm. The days required for biodegradation of this film were 30 days.

Example 2.6

Pellets were formed by blending 40 parts by weight of calcium carbonate (made by Maruo Calcium Co., trade name: Super SS, mean grain size: 2.2 μm) relative to 100 parts by weight of a polyesteramide resin (melting point: 172°C), produced by polymerization of adipic acid, 1,4-butanediol and hexamethylene diamine, comprising 80% by weight of an aliphatic ester unit and 20% by weight of an aliphatic amide unit. A film with a thickness of 30 μm was formed from the pellet using a T-die type extruder equipped with a cylinder with a diameter of 30 mm. The days required for biodegradation of this film were 33 days.

Comparative Example 2.2

A film was formed by the same method as used in Example 2.1, except that calcium carbonate used in Example 2.6 was not blended at all. The days required for biodegradation of this film were 52 days.

Example 3.1

Into 100 parts by weight of the resin component comprising the polyesteramide resin (melting point 137°C), produced by polymerization of adipic acid, 1,4-butanediol and ε-caprolactam, comprising 50% by weight of an aliphatic ester unit and 50% by weight of an aliphatic amide unit0.1 parts by weight of a hindered phenol based antioxidant (made by Ashahi Electrochemical Co., trade name AO-60), 0.1 parts by weight of a phosphite ester based compound (made by Ashahi Electrochemical Co., trade name "2112"), 0.3 parts by weight of a hindered amine based light stabilizer (made by Ciba Speciality Chemical Co., trade name LS-944), 0.1 parts by weight of a triazole based UV absorbing agent (made by Kyodo Pharmaceuticals Co., trade name VS-550), and 0.1 parts by weight of a fluorinated surfactant (made by Daikin Industries, Co.) as a clod preventive agent and fog preventive agent comprising 1 part by weight of diglycerine distealate and 1 parts by weight of sorbitan monostealate

were blended. A biodegradable agricultural film with a thickness of 100 μm was produced with an inflation molding machine.

This film was evaluated as follows: haze; 13%, cloud preventive property; O, tensile strength; 680 kg/cm^2, extension; 500%, weather resistance; O, biodegradability; O

Example 3.2

A biodegradable agricultural film was produced by the same method as in Example 3.1, except that the resin component comprises 95 parts by weight of the poly-esteramide resin used in Example 3.1 and 5 parts by weight of an aliphatic polyester resin (made by Showa Polymer Industry Co., trade name Bionole 3001).

This film was evaluated as follows: haze; 15%, cloud preventive property; O, tensile strength; 710 kg/cm^2, extension; 480%, weather resistance; O, biodegradability; O

Example 3.3

A biodegradable agricultural film was produced by the same method as in Example 3.1, except that the resin component comprises 90 parts by weight of the poly-esteramide resin used in Example 1 and 10 parts by weight of an aliphatic polyester resin (made by Showa Polymer Industry Co., trade name Bionole 3001).

This film was evaluated as follows: haze; 17%, cloud preventive property; O, tensile strength; 740 kg/cm^2, extension; 450%, weather resistance; O, biodegradability; O

Comparative Example 3.1

A commercially available agricultural soft polyvinyl chloride film with a thickness of 100 μ (made by C.I. Chemicals Co., trade name C.I. agricultural polyvinyl chloride Sky Eight) was evaluated, obtaining the following results:

haze; 15%, cloud preventive property; O, tensile strength; 250 kg/cm^2, extension; 320%, weather resistance; O, biodegradability; ×

<u>Comparative Example 3.2</u>

A commercially available agricultural polyolefine film with a thickness of 100 μm (made by C.I. Chemicals Co., trade name C.I. agricultural polyvinyl chloride Sky Coat) was evaluated, obtaining the following results:
haze; 15.5%, cloud preventive property; O, tensile strength; 224 kg/cm^2, extension; 550%, weather resistance; O, biodegradability; ×

<u>Comparative Example 3.3</u>

A biodegradable agricultural film was produced by the same method as in Example 3.1, except that 100 parts by weight of the aliphatic polyester resin (made bay Showa Polymer Industry Co., trade name Bionole 3001) was used as the resin component.

The film was evaluated as follows: haze; 68%, cloud preventive property; ×, tensile strength; 610 kg/cm^2, extension; 840%, weather resistance; O, biodegradability; O

<u>Example 3.5</u>

A biodegradable agricultural film was produced by the same method as in Example 1, except that 100 parts by weight of a polyesteramide resin (melting point 116°C), produced by polymerization of adipic acid, 1,4-butanediol and ε-caprolactam, comprising 70% by weight of an aliphatic amide unit and 30% by weight of an aliphatic polyester unit was used.

The film was evaluated as: haze; 15%, cloud preventive property; O, tensile strength; 660 kg/cm^2, extension; 520%, weather resistance; O, biodegradability; O

Example 3.6

A biodegradable agricultural film was produced by the same method as in Example 3.1, except that 90 parts by weight of the polyesteramide resin (melting point 116°C) used in Example 3.5 and 10 parts by weight of the aliphatic polyester resin (made by Showa Polymer Industry Co., trade name Bionole 3001) were used as the resin component.

The film was evaluated as: haze; 19%, cloud preventive property; O, tensile strength; 710 kg/cm^2, extension; 460%, weather resistance; O, biodegradability; O

Example 3.7

A biodegradable agricultural film was produced by the same method as in Example 1, except that 100 parts by weight of a polyesteramide resin (melting point 172°C), produced by polymerization of adipic acid, 1,4-butanediol and hexamethylene diamine, comprising 80% by weight of an aliphatic ester unit and 20% by weight of an aliphatic amide unit was used.

The film was evaluated as: haze; 19%, cloud preventive property; O, tensile strength; 820 kg/cm^2, extension; 400%, weather resistance; O, biodegradability; O

Example 4.1

Additives such as 0.1 parts by weight of the hindered phenol based antioxidant (made by Asahi Electrochemical Co., trade name AO-80), 0.1 parts by weight of the phosphite ester based compound (made by Asahi Electrochemical Co., trade name "2112"), 0.02 parts by weight of the hindered amine based light stabilizer (made by Ciba Speciality Chemicals Co., trade name LS-944), and 30 parts by weight of calcium carbonate (made by Maruo Calcium Co., mean grain size 2.2 µm) were blended into 100 parts by weight of the resin component composed of a polyesteramide resin

(melting point 137°C), produced by polymerization of adipic acid, 1,4-butanediol and ε-caprolactam, comprising 50% by weight of the aliphatic ester unit and 50% by weight of the aliphatic amide unit. An arvicultural multi-film of the biodegradable resin with a thickness of 30 μm was produced with an inflation-molding machine.

This multi-film was evaluated as: tensile strength; 310 kg/cm^2, extension; 370%, perpendicular tear strength; 120 kg/cm^2, biodegradability; ◎

Example 4.2

An arvicultural multi-film of the biodegradable resin was produced by the same method as in Example 4.1, except that 3 parts by weight of carbon black was further added to the composition used in Example 4.1.

This multi-film was evaluated as: tensile strength; 320 kg/cm^2, extension; 360%, perpendicular tear strength; 110 kg/cm^2, biodegradability; ◎

Example 4.3

An arvicultural multi-film of the biodegradable resin was produced by the same method as in Example 4.1, except that the resin component comprising 95 parts by weight of the polyesteramide resin used in Example 4.1 and 5 parts by weight of an aliphatic polyester resin (made by Showa Polymer Industry Co., trade name Bionole 3001) was used.

This multi-film was evaluated as: tensile strength; 380 kg/cm^2, extension; 330%, perpendicular tear strength; 130 kg/cm^2, biodegradability; ◎

Example 4.4

Additives such as 0.1 parts by weight of the hindered phenol based antioxidant (made by Asahi Electrochemical Co., trade name AO-80), 0.1 parts by weight of the phosphite ester based compound (made by Asahi Electrochemical Co., trade name "2112"), 30 parts by weight of calcium carbonate (made by Maruo Calcium Co., mean grain size 2.2 μm) as a filler and 0.8 parts by weight of a green pigment (made by Toyo Inc Co., trade name Lionol Green Y-101) were blended into 100 parts by weight of the polyesteramide resin used in Example 1. An agricultural multi-film of the biodegradable resin with a thickness of 30 μm was produced with an inflation-molding machine.

This multi-film was evaluated as: tensile strength; 320 kg/cm^2, extension; 370%, perpendicular tear strength; 120 kg/cm^2, biodegradability; ◎

Comparative Example 4.1

A commercially available black poly multi-film with a thickness of 100 μm was evaluated, obtaining the following results: tensile strength; 350 kg/cm^2, extension; 410%, perpendicular tear strength; 160 kg/cm^2, biodegradability; ×

Example 4.5

An arvicultural multi-film of the biodegradable resin was produced by the same method as in Example 4.1, except that a polyesteramide resin (melting point 116 °C), produced by polymerization of adipic acid, 1,4-butanediol and ε-caprolactam, comprising 30% by weight of the aliphatic resin unit and 70% by weight of the aliphatic amide unit was used as the resin component.

This multi-film was evaluated as: tensile strength; 300 kg/cm^2, extension; 390%, perpendicular tear strength; 130 kg/cm^2, biodegradability; ◎

Example 4.6

An agricultural multi-film was produced by the same method as in Example 4.1, except that 90 parts by weight of the aliphatic polyesteramide resin (melting point 172°C), produced by polymerization of adipic acid, 1,4-butanediol and hexamethylene diamine, comprising 80% by weight of the aliphatic ester unit and 20% by weight of the aliphatic amide unit, and 10 parts by weight of the aliphatic polyester resin (made by Showa Polymer Industry Co., trade name Bionole 3001) were used as the resin component.

This multi-film was evaluated as: tensile strength; 380 kg/cm^2, extension; 310%, perpendicular tear strength; 110 kg/cm^2, biodegradability; ◎

Example 5.1

A resin component composed of 80 parts by weight of the polyesteramide resin, prepared by polymerization of adipic acid, 1,4-butane diol and ε-caprolactam, comprising 50% of an aliphatic ester unit and 50% of an aliphatic amide unit, and 20 parts by weight of the aliphatic polyester resin (made by Showa Polymer Industry Co., trade name Bionole 3001) was used as the layer (B), and a resin component composed of 100 parts by weight of the aliphatic polyester resin (made by Showa Polymer Industry Co., trade name Bionole 3001) was used as the layer (B). Into 100 parts by weight of the resin component for each layer, 0.1 parts by weight of a hindered phenol based antioxidant (made by Asahi Electrochemical Co., trade name AO-80), 0.1 parts by weight of a phosphite ester based compound (made by Asahi Electrochemical Co., trade name "2112"), and 0.2 parts by weight of a hindered amine based light stabilizer (made by Ciba Speciality Chemical Co., trade name LS-944) were blended. A biodegradable multilayer film having a construction of (B)/(A)/(B) with a layer ratio of 10/80/10 and a thickness of 100 μm was formed using a three-layer inflation molding machine.

This film was evaluated as: water resistance; ◎, haze; 19%, flexibility; 41 kg/mm^2, biodegradability; O

<u>Example 5.2</u>

A resin component composed of 100 parts by weight of the polyesteramide resin, prepared by polymerization of adipic acid, 1,4-butane diol and ε-caprolactam, comprising 50% of an aliphatic ester unit and 50% of an aliphatic amide unit was used as the layer (B), and a resin component composed of 80 parts by weight of the aliphatic polyester resin (made by Showa polymer Industry Co., trade name Bionole 3001) and 20 parts by weight of the polyesteramide resin was used as the layer (B). Into 100 parts by weight of the resin component for each layer, 0.1 parts by weight of a hindered phenol based antioxidant (made by Asahi Electrochemical Co., trade name AO-80), 0.1 parts by weight of a phosphite ester based compound and 0.2 parts by weight of a hindered amine based light stabilizer (made by Ciba Speciality Chemical Co., trade name LS-944) were blended. A biodegradable multilayer film having a layer construction of (B)/(A)/(B) with a layer ratio of 10/80/10 and a thickness of 100 μm was formed using a three-layer inflation molding machine.

This film was evaluated as: water resistance; O, haze; 21%, flexibility; 39 kg/mm^2, biodegradability; ◎

<u>Example 5.3</u>

A resin component composed of 70 parts by weight of the polyesteramide resin, prepared by polymerization of adipic acid, 1,4-butane diol and ε-caprolactam, comprising 30% of an aliphatic ester unit and 70% of an aliphatic amide unit was used as the layer (A), and a resin component composed of 100 parts by weight of the aliphatic polyester resin (made by Showa polymer Industry Co., trade name Bionole 3001) was used as the layer (B). Into 100 parts by weight of the resin component for each

layer, 0.1 parts by weight of a hindered phenol based antioxidant (made by Asahi Electrochemical Co., trade name AO-80), 0.1 parts by weight of a phosphite ester based compound (made by Asahi Electrochemical Co., trade name "2112"), 0.2 parts by weight of a hindered amine based light stabilizer (made by Ciba Speciality Chemical Co., trade name LS-944), and 30 parts by weight of calcium carbonate (made by Maruo Calcium Co., mean grain size: 2,2 μm) as an additive were blended. A biodegradable multilayer film having a layer construction of (B)/(A)/(B) with a layer ratio of 15/70/15 and a thickness of 100 μm was formed using a three-layer inflation molding machine.

This film was evaluated as: water resistance; ◎, haze; 21%, flexibility; 46 kg/mm^2, biodegradability; O

Comparative Example 5.1

Into 100 parts by weight of the polyesteramide resin, prepared by polymerization of adipic acid, 1,4-butanediol and ε-caprolactam, comprising 50% of an aliphatic ester unit and 50% of an aliphatic amide unit, 0.1 parts by weight of the hindered phenol based antioxidant (made by Asahi Electrochemical Co., trade name AO-80), 0.1 parts by weight of a phosphite ester based compound (made by Asahi Electrochemical Co., trade name "2112"), and 0.2 pars by weight of the hindered amine based light stabilizer (made by Ciba Speciality Chemical Co., trade name LS-944) were blended. A single layer biodegradable film with a thickness of 100 μm was formed from this composition using a single layer inflation molding machine.

This film was evaluated as: water resistance; Δ, haze; 14%, flexibility; 36 kg/mm^2, biodegradability; ◎

<u>Comparative Example 5.2</u>

Into 100 parts by weight of the aliphatic polyesteramide resin (made by Showa Polymer Industry Co. trade name Bionole 3001), 0.1 parts by weight of the hindered phenol based antioxidant (made by Asahi Electrochemical Co., trade name AO-80), 0.1 parts by weight of a phosphite ester based compound (made by Asahi Electrochemical Co., trade name "2112"), and 0.2 pars by weight of the hindered amine based light stabilizer (made by Ciba Speciality Chemical Co., trade name LS-944) were blended. A single layer biodegradable film with a thickness of 100 μm was formed from this composition using a single layer inflation molding machine.

This film was evaluated as: water resistance; ◎, haze; 68%, flexibility; 800 kg/mm^2, biodegradability; O, especially showing poor transparency and flexibility.

<u>Example 5.4</u>

A resin component composed of 80 parts by weight of the aliphatic polyesteramide resin, prepared by polymerization of adipic acid, 1,4-butane diol and hexamethylene diamine, comprising 80% of the aliphatic ester unit and 20% of the aliphatic amide unit, and 20 parts by weight of an aliphatic polyester resin (made by Showa Polymer Industry Co., trade name Bionole 3001) was used as the layer (A), and a resin component composed of 100 parts by weight of the aliphatic polyester resin (made by Showa polymer Industry Co., trade name Bionole 3001) was used as the layer (B). Into 100 parts by weight of the resin component for each layer, 0.1 parts by weight of a hindered phenol based antioxidant (made by Asahi Electrochemical Co., trade name AO-80), 0.1 parts by weight of a phosphite ester based compound (made by Asahi Electrochemical Co., trade name "2112"), 0.2 parts by weight of a hindered amine based light stabilizer (made by Ciba Speciality Chemical Co., trade name LS-944), and 30 parts by weight of calcium carbonate (made by Maruo Calcium Co., mean grain size: 2,2 μm) as an additive were blended. A biodegradable multilayer

film having a layer construction of (B)/(A)/(B) with a layer ratio of 15/70/15 and a thickness of 100 μm was formed using a three-layer inflation molding machine.

This film was evaluated as: water resistance; ◎, haze; 24%, flexibility; 65 kg/mm^2, biodegradability; O

Example 6.1

A resin component composed of 100 parts by weight of an aliphatic polyester resin (made by Showa Polymer Industry Co., trade name Bionole 3001) was used as the layer (A); a resin component composed of 70 parts by weight of a polyesteramide rsin, prepared by polymerization of adipic acid, 1,4-bitanediol and ε-caprolactam, comprising 50% of an aliphatic ester unit and 50% of an aliphatic amide unit, and 30 parts by weight of the aliphatic polyester resin described above was used as the layer (B); and a resin component composed of 80 parts by weight of the aliphatic polyester resin and 20 parts by weight of the polyesteramide resin was used as the layer (C). To 100 parts by weight of the resin component for each layer, 0.1 parts by weight of a hindered phenol based anti-oxidant (made by Asahi Electrochemical Co., trade name AO-80), 0.1 parts by weight of a phosphite ester based compound (made by Asahi Electrochemical Co., trade name "2112"), and 0.3 parts by weight of a hindered amine based light stabilizer (made by Ciba Speciality Chemical Co., trade name LS-944) were blended. A cloud preventive agent comprising 1 part by weight of diglycerine distearate and 1 parts by weight of sorbitan monostearate, and a fog preventive agent comprising 0.1 parts by weight of a fluorinated surfactant (made by Daikin Industries Co., trade name DS-403) were blended to the layer (B); and a cloud preventive agent comprising 0.7 parts by weight of diglycerine distearate and 0.7 parts by weight of sorbitan monostearate, and a fog preventive agent comprising 0.1 parts by weight of a fluorinated surfactant (made by Daikin Industries Co., trade name DS-403) were blended to the layer (C).

A biodegradable multilayer agricultural film with a construction of (A)/(B)/(C), a layer ratio of 20/60/20 and a thickness of 100 μm was produced using a three-layer inflation molding machine.

This film was evaluated as; haze: 23%, cloud preventive property: O, water resistance: O, acceleration of weather resistance: O, cloud resistant blocking property: O

Example 6.2

A biodegradable multilayer agricultural film was produced by the same method as in Example 6.1, except that the resin component in the layer (C) was replaced with 70 parts by weight of the aliphatic polyester resin and 30 parts by weight of the polyesteramide resin.

This film was evaluated as; haze: 21%, cloud preventive property: ◎, water resistance: O, acceleration of weather resistance: O, heat resistant blocking property: O

Example 6.3

A resin component composed of 100 parts by weight of an aliphatic polyester resin (made by Showa Polymer Industry Co., trade name Bionole 3001) was used as the layer (A); a resin component composed of 80 parts by weight of a polyesteramide resin, prepared by polymerization of adipic acid, 1,4-butanediol and ε-caprolactam, comprising 30% of an aliphatic ester unit and 70% of an aliphatic amide unit, and 20 parts by weight of the aliphatic polyester resin described above was used as the layer (B); and a resin component composed of 70 parts by weight of the aliphatic polyester resin and 30 parts by weight of the polyesteramide resin was used as the layer (C). To 100 parts by weight of the resin component for each layer, 0.1 parts by weight of a hindered phenol based anti-oxidant (made by Asahi Electrochemical Co., trade name AO-80), 0.1 parts by weight of a phosphite ester based compound (made by Asahi Electrochemical Co., trade name "2112"), and 0.3 parts by weight of a hin-

dered amine based light stabilizer (made by Asahi Electrochemical Co., trade name LA-63) were blended. A cloud preventive agent comprising 1 part by weight of diglycerine distearate and 1 parts by weight of sorbitan monostearate, and a fog preventive agent comprising 0.1 parts by weight of a fluorinated surfactant (made by Daikin Industries Co., trade name DS-403) were blended to the layer (B); and a cloud preventive agent comprising 0.7 parts by weight of diglyceride stearate and 0.7 parts by weight of sorbitan monostearate, and a fog preventive agent comprising 0.1 parts by weight of a fluorinated surfactant (made by Daikin Industries Co., trade name DS-403) were blended to the layer (C).

A biodegradable multilayer agricultural film with a construction of (A)/(B)/(C), a layer ratio of 15/70/15 and a thickness of 100 μm was produced using a three-layer inflation molding machine.

This film was evaluated as; haze: 22%, cloud preventive property: ◎, water resistance: O, acceleration of weather resistance: O, heat resistant blocking property: O

Example 6.4

A resin component composed of 100 parts by weight of an aliphatic polyester resin (made by Showa Polymer Industry Co., trade name Bionole 3001) was used as the layer (A); a resin component composed of 80 parts by weight of a polyesteramide resin, prepared by polymerization of adipic acid, 1,4-butanediol and hexamethylene diamine, comprising 80% of an aliphatic ester unit and 20% of an aliphatic amide unit, and 20 parts by weight of the aliphatic polyester resin described above was used as the layer (B); and a resin component composed of 70 parts by weight of the aliphatic polyester resin and 30 parts by weight of the polyesteramide resin was used as the layer (C). To 100 parts by weight of the resin component for each layer, 0.1 parts by weight of a hindered phenol based anti-oxidant (made by Asahi Electrochemical Co., trade name AO-80), 0.1 parts by weight of a phosphite ester based compound (made by Asahi Electrochemical Co., trade name "2112"), and 0.3 parts by weight of

a hindered amine based light stabilizer (made by Asahi Electrochemical Co., trade name, trade name LA-63) were blended. A cloud preventive agent comprising 1 part by weight of diglycerine distearate and 1 parts by weight of sorbitan monostearate, and a fog preventive agent comprising 0.1 parts by weight of a fluorinated surfactant (made by Daikin Industries Co., trade name DS-403) were blended to the layer (B); and a cloud preventive agent comprising 0.7 parts by weight of diglyceride stearate and 0.7 parts by weight of sorbitan monostearate, and a fog preventive agent comprising 0.1 parts by weight of a fluorinated surfactant (made by Daikin Industries Co., trade name DS-403) were blended to the layer (C).

A biodegradable multilayer agricultural film with a construction of (A)/(B)/(C), a layer ratio of 15/70/15 and a thickness of 100 μm was produced using a three-layer inflation molding machine.

This film was evaluated as; haze: 22%, cloud preventive property: ◎, water resistance: O, acceleration of weather resistance: O, heat resistant blocking property: O

<u>Comparative Example 6.1</u>

To 100 parts by weight of a polyesteramide resin, prepared by polymerization of adipic acid, 1,4-butanediol and ε-caprolactam, comprising 50% of the aliphatic ester unit and 50% of the aliphatic amide unit, 0.1 parts by weight of a hindered phenol based anti-oxidant (made by Asahi Electrochemical Co., trade name AO-80), 0.1 parts by weight of a phosphite ester based compound (made by Asahi Electrochemical Co., trade name "2112"), and 0.3 parts by weight of a hindered amine based light stabilizer (made by Ciba Speciality Chemical Co., trade name LS-944) were blended, as well as a cloud preventive agent comprising 1 part by weight of diglycerine distearate and 1 part by weight of sorbitan monostearate, and a fog preventive agent comprising 0.1 parts by weight of a fluorinated surfactant (made by Daikin Industries Co., trade name DS-403). A biodegradable mono-layer agricultural film a thickness of 100 μm was produced using a mono-layer inflation molding machine.

This film was evaluated as; haze: 14%, cloud preventive property: ◎, water resistance: Δ, acceleration of weather resistance: O, heat resistant blocking property: Δ

Comparative Example 6.2

To 100 parts by weight of the aliphatic polyester resin (made by Showa Polymer Industry Co., trade name Bionole 3001), 0.1 parts by weight of a hindered phenol based anti-oxidant (made by Asahi Electrochemical Co., trade name AO-80), 0.1 parts by weight of a phosphite ester based compound (made by Asahi Electrochemical Co., trade name "2112"), and 0.3 parts by weight of a hindered amine based light stabilizer (made by Ciba Speciality Chemical Co., trade name LS-944) were blended, as well as a cloud preventive agent comprising 1 part by weight of diglycerine distearate and 1 part by weight of sorbitan monostearate, and a fog preventive agent comprising 0.1 parts by weight of a fluorinated surfactant (made by Daikin Industries Co., trade name DS-403). A biodegradable mono-layer agricultural film a thickness of 100 μm was produced using a mono-layer inflation molding machine.

This film was evaluated as; haze: 68%, cloud preventive property: ×, water resistance: ◎, acceleration of weather resistance: O, heat resistant blocking property: O

Example 7.1

Additives such as 0.1 parts by weight of a hindered phenol based antioxidant (made by Asahi Electrochemical Co., trade name AO-80), 0.1 parts by weight of a phosphite ester based compound (made by Asahi Electrochemical Co., trade name "2112"), 0.2 parts by weight of a hindered amine based light stabilizer (made by Ciba Speciality Chemical Co., trade name LS-944), and 30 parts by weight of calcium carbonate (made by Maruo Calcium Co, mean grain size: 2.2 μm) as a filler were blended into 100 parts by weight of a resin component composed of a polyesteramide resin (melting point 137 °C), prepared by polymerization of adipic acid, 1,4-butane-

diol and ε-caprolactam, comprising 50% by weight of an aliphatic ester unit and 50% by weight of an aliphatic amide unit. A multi-film of a biodegradable resin for use in rice fields with a thickness of 30 μm was produced using an inflation molding machine.

It was possible to plant young rice plants into a rice field while covering the rice field with a multi-film of the biodegradable resin for use in rice fields by piercing the multi-film with a rice planting machine.

The multi-film was evaluated as; tensile strength (moist film): 250 kg/cm^2, extension (moist film): 390%, flexibility (moist film): 18 kg/mm^2, biodegradability ◎

Example 7.2

A multi-film of a biodegradable resin for use in rice fields was produced by the same method as in Example 7.1, except that 3 parts by weight of carbon black was used in addition to the additive used in Example 7.1.

It was possible to plant young rice plants into a rice field while covering the rice field with a multi-film of the biodegradable resin for use in rice fields by piercing the multi-film with a rice planting machine.

The multi-film was evaluated as; tensile strength (moist film): 260 kg/cm^2, extension (moist film): 370%, flexibility (moist film): 19 kg/mm^2, biodegradability ◎

Example 7.3

A multi-film of a biodegradable resin for use in rice fields was produced by the same method as in Example 7.1, except that the proportion of blending of the polyester amide resin and aliphatic polyester resin used in Example 7.1 were changed to 95 parts by weight and 5 parts by weight, respectively.

It was possible to plant young rice plants into a rice field while covering the rice field with a multi-film of the biodegradable resin for use in rice fields by piercing the multi-film with a rice planting machine.

The multi-film was evaluated as; tensile strength (moist film): 330 kg/cm^2, extension (moist film): 340%, flexibility (moist film): 22 kg/mm^2, biodegradability ◎.

Example 7.4

Additives such as 0.1 parts by weight of a hindered phenol based antioxidant (made by Asahi Electrochemical Co., trade name AO-80), 0.1 parts by weight of a phosphite ester based compound (made by Asahi Electrochemical Co., trade name "2112"), 30 parts by weight of calcium carbonate (made by Maruo Calcium Co, mean grain size: 2.2 μm) as a filler, and 0.8 parts by weight of a green pigment (made by Toyo Ink Co., trade name Lionol Green Y-101) as a colorant were blended into 100 parts by weight of the polyesteramide resin used in Example 7.1. A multi-film of a biodegradable resin for use in rice fields with a thickness of 30 μm was produced using an inflation molding machine.

It was possible to plant young rice plants into a rice field while covering the rice field with a multi-film of the biodegradable resin for use in rice fields by piercing the multi-film with a rice planting machine.

The multi-film was evaluated as; tensile strength (moist film): 260 kg/cm^2, extension (moist film): 380%, flexibility (moist film): 19 kg/mm^2, biodegradability ◎.

Comparative Example 7.1

A commercially available black poly-multifilm with a thickness of 30 μm was evaluated as; tensile strength (moist film): 340 kg/cm^2, extension (moist film): 400%, flexibility (moist film): 14 kg/mm^2, biodegradability ×

Comparative Example 7.2

Additives such as 0.1 parts by weight of a hindered phenol based antioxidant (made by Asahi Electrochemical Co., trade name AO-80), 0.1 parts by weight of a phosphite ester based compound (made by Asahi Electrochemical Co., trade name "2112"), 0.2 parts by weight of a hindered amine based light stabilizer (made by Ciba Speciality Chemical Co., trade name LS-944), and 30 parts by weight of calcium carbonate (made by Maruo Calcium Co, mean grain size: 2.2 μm) as a filler were blended into 100 parts by weight of the aliphatic polyester resin (made by Showa Polymer Industry Co., trade name Bionole 3001). A multi-film of a biodegradable resin for use in rice fields with a thickness of 30 μm was produced using an inflation molding machine.

Young rice plants were planted into a rice field using a planting machine while covering the rice field with the multi-film of the biodegradable resin. However, the film was so hard that rice planting work was difficult.

Also, the film has so poor flexibility that it could not make close contact to the surface of the soil, causing such troubles as the film get rolled up by wind.

The multi-film was evaluated as; tensile strength (moist film): 590 kg/cm^2, extension (moist film): 850%, flexibility (moist film): 76 kg/mm^2, biodegradability O.

Example 7.5

A multi-film of a biodegradable resin for use in rice fields was produced by the same method as in Example 7.1, except that a polyesteramide resin (melting point: 116°C), prepared by polymerization of adipic acid, 1,4-butanediol and ε-caprolactam, comprising 30% by weight of the aliphatic ester unit and 70% by weight of the aliphatic amide unit was used as the resin component.

It was possible to plant young rice plants into a rice field while covering the rice field with a multi-film of the biodegradable resin for use in rice fields by piercing the multi-film with a rice planting machine.

The multi-film was evaluated as; tensile strength (moist film): 240 kg/cm^2, extension (moist film): 400%, flexibility (moist film): 19 kg/mm^2, biodegradability ◎.

Example 7.6

A multi-film of a biodegradable resin for use in rice fields was produced by the same method as in Example 7.1, except that 90 parts by weight of a polyesteramide resin (melting point: 172°C), prepared by polymerization of adipic acid, 1,4-butanediol and hexamethylene diamine, comprising 80% by weight of the aliphatic ester unit and 20% by weight of the aliphatic amide unit, and 10 parts by weight of a aliphatic polyester resin (made by Showa Polymer Industry Co., trade name Bionole 3001) were used as the resin components.

It was possible to plant young rice plants into a rice field while covering the rice field with a multi-film of the biodegradable resin for use in rice fields by piercing the multi-film with a rice planting machine.

The multi-film was evaluated as; tensile strength (moist film): 320 kg/cm^2, extension (moist film): 320%, flexibility (moist film): 22 kg/mm^2, biodegradability ◎.

Claims

1. Biodegradable resin composition comprising at least one stabilizing additive selected from the groups of antioxidants (a), of radical scavenging light stabilizers (b), UV and visible light absorbing compounds (c), and quenchers of photochemical excited states (d), additional additives like anti-clouding agents (e) and anti-fogging agents (f) and comprising at least one biodegradable polymer selected from the group consisting of aliphatic (co)polyesters, aromatic-aliphatic (co)polyesters, aliphatic polycarbonates, aromatic-aliphatic polycarbonates, aliphatic polyester-urethanes, partly aromatic polyester-urethanes, aliphatic polyesteramides, aliphatic-aromatic polyesteramides, polyetheresteramides, polysaccharide esters, polysaccharide ether esters such as cellulose or starch derivatives or copolymers and/or mixtures thereof.

2. Resin composition according to claim 1 comprising fillers.

3. A resin composition according to claim 1 or 2, wherein the polymer is chosen from a polymer of the group consisting of:

aliphatic or partly aromatic polyesters from

A) aliphatic bifunctional alcohols, and/or optionaly cycloaliphatic bifunctional alcohols, and/or, instead of some or all of the diols, monomeric or oligomeric polyols based on ethylene glycol, propylene glycol or tetrahydrofuran or copolymers thereof having molecular weights of up to 8,000, and/or optionally small amounts of branched bifunctional alcohols, and additionally optionally small amounts of alcohols of higher functionality, and from aliphatic bifunctional acids, and/or optionally aromatic bifunctional acids and additionally optionally small amounts of acids of higher functionality or

B) building blocks with acid and alcohol functional groups or derivatives thereof, or a mixture and/or copolymer of A and B, the aromatic acids making up a content of not more than 50 wt.%, based on all the acids;

aliphatic or partly aromatic polyester-urethanes from

C) aliphatic bifunctional alcohols, and/or optionally cycloaliphatic bifunctional alcohols, and/or, instead of some or all of the diols, monomeric or oligomeric polyols based on ethylene glycol, propylene glycol or tetrahydrofuran or copolymers thereof having molecular weights of up to 4,000, and/or small amounts of branched bifunctional alcohols, and additionally optionally small amounts of alcohols of higher functionality, and from aliphatic bifunctional acids, and/or optionally aromatic bifunctional acids and additonally optionally small amounts of acids of higher functionality, or

D) building blocks with acid and alcohol functional groups, or derivatives thereof, or a mixture and/or copolymer of C and D,

the aromatic acids making up a content of not more than 50 wt.%,

based on all the acids;

the reaction product of C and/or D and

E) aliphatic and/or cycloaliphatic bifunctional isocyanates and additionally optionally isocyanates of higher functionality, optionally additionally with linear and/or branched and/or cycloaliphatic bifunctional alcohols and/or alcohols of higher functionality, and/or op-

tionally additionally with linear and/or branched and/or cycloaliphatic bifunctional amines and/or amino alcohols and/or amines and/or amino alcohols of higher functionality and/or optionally further modified amines or alcohols,

the ester content C) and/or D) being at least 75 wt.%, based on the sum of C), D) and E);

aliphatic or aliphatic-aromatic polyester-carbonates from

F) aliphatic bifunctional alcohols, and/or optionally cycloaliphatic bifunctional alcohols, and/or, instead of some or all of the diols, monomeric or oligomeric polyols based on ethylene glycol, propylene glycol or tetrahydrofuran or copolymers thereof having molecular weights of up to 4,000, and/or optionally small amounts of branched bifunctional alcohols, and additionally optionally small amounts of alcohols of higher functionality, and from aliphatic bifunctional acids, and/or optionally aromatic bifunctional acids and additionally optionally small amounts of acids of higher functionality or

G) building blocks with acid and alcohol functional groups or derivatives thereof, or a mixture and/or copolymer of F) and G),

the aromatic acids making up a content of not more than 50 wt.%, based on all the acids;

with

H) a carbonate content which is prepared from aromatic bifunctional phenols and carbonate donors, or a carbonate content which is pre-

pared from aliphatic carbonic acid esters or derivatives thereof and carbonate donors

the ester content F) and/or G) being at least 70 wt.%, based on the sum of F), G) and H);

aliphatic or partly aromatic polyester-amides or polyether-ester-amides from

I) aliphatic bifunctional alcohols, and/or optionally cycloaliphatic bifunctional alcohols, and/or, instead of some or all of the diols, monomeric or oligomeric polyols based on ethylene glycol, propylene glycol or tetrahydrofuran or copolymers thereof having molecular weights of up to 10,000, and/or optionally small amounts of branched bifunctional alcohols, and additionally small amounts of alcohols of higher functionality, and from aliphatic bifunctional acids, and/or optionally aromatic bifunctional acids and additionally optionally small amounts of acids of higher functionality or

K) building blocks with acid and alkohol functional groups or derivatives thereof,

or a mixture and/or a copolymer of I) and K), the aromatic acids

and

L) an amide content from aliphatic and/or cycloaliphatic bifunctional and/or optionally small amounts of branched bifunctional amines, and additionally optionally small amounts of amines of higher functionality, and from linear and/or cycloaliphatic bifunctional acids, and/or optionally small amounts of branched bifunctional and/or op-

tionally aromatic bifunctional acids and additionally optionally small amounts of acids of higher functionality, or

M) an amide content of building blocks with acid and amine functional groups,

or a mixture of L) and M) as the amide content, the ester content I) and/or K) being at least 30 wt.%, based on the sum of I), K), L) and M).

4. Resin composition according to claims 1-3, wherein the polymer is selected from aliphatic (co)polyesters, aromatic-aliphatic (co)polyesters, polyether-esteramides and/or aliphatic polyesteramides.

4a. Resin composition according to claim 1-4, wherein

0-1,0 parts by weight of c),
0-1,2 parts by weight of a), 0-1,0 parts by weight of b) and polymer add up to 100 parts by weight, optionally d), e) and f) may be added.

5. Resin composition according to claims 1 to 4a, wherein the antioxidant (a) is at least one selected from the group of hindered phenol based compounds (a1) and/or phosphite and/or phosphonite ester based compounds (a2) and/or sulfur containing synergists in antioxidation (a3) and/or the light stabilizer (b) is at least one selected from the hindered amine based light stabilizers, preferably a high molecular weight oligomer or polymer compound comprising at least two repeating units and/or the UV-absorber (c) is at least one selected from the group of benzophenones, benzotriazoles, benzylidene malonates, oxanilides, benzoaxazinones or triazines; the quencher (d) is selected from the metal organic compounds.

6. Use of resin composition according claim for molded articles, preferred films and sheets.

7. Molded articles, produced according to 1-6.

8. Use of the resin compositions according to claims 1 to 5 for films characterized that at least one of the additves (a) to (f) are blended into the resin composition and the biodegradable resin composition is prepared by blending relative to 100 parts by weight of the composition 0.03 to 1.2 parts by weight of an antioxidant (a) and/or the resin composition comprises a hindered amine based light stabilizer (b) in an amount of 0.03 to 1.0 parts by weight, relative to 100 parts by weight of the composition and/or comprises an UV-absorber (c) in an amount of 0.01 to 1.0 parts by weight, relative to 100 parts by weight of the composition.

9. Film according to claim 8.

10. Film according to claim 9 with a thickness in the range of 15 to 1000 μm.

11. Use of resin composition according to claims 1 to 5 for agricultural films and sheets of improved weatherability that are transparent wherein the preferred composition of the biodegradable resin comprises a blend of a biodegradable polyesteramide, preferably an aliphatic polyesteramide, and a biodegradable polyester, preferably an aliphatic polyester.

12. Use of resin composition according to claim 11 for transparent agricultural films and sheets wherein the resin composition comprises 60 to 95 parts by weight of the polyesteramide and 5 to 40 parts by weight of the aliphatic polyester and the amount of the hindered amine based light stabilizer being added to this composition is in the range of 0.03 to 1.0 parts by weight, relative to 100 parts by weight of the composition.

13. Agricultural transparent films according to claims 11 and/or 12.

14. Agricultural films according to claim 14 with a film thickness of 20 to 200 µm.

15. Use of resin compositions according to claims 1 to 5 for multi-layer films and sheets wherein the resin composition comprises 60 to 100 parts by weight of the polyesteramide resin and 0 to 40 parts by weight of the aliphatic polyester resin and the ratio of the hindered phenol based antioxidant (a1) and/or phosphite and/or phosphonite ester based antioxidant (a2) is in the range of 0.01 to 1.0 parts by weight, relative to 100 parts by weight of the resin composition and the ratio of the hindered amine based light stabilizer (b) is in the range of 0.01 to 1.0 parts by weight, relative to 100 parts by weight of the resin composition and the additives blended in a range of 0.5 to 50 parts by weight, relative to 100 parts by weight of the resin composition.

16. Multilayer films and sheets according to claim 15.

17. Multilayer films and sheets according to claim 16, characterized that the film thickness is in the range of 15 to 100 µm.

18. Use of resins according to claims 1 to 17 for multilayer films provided with a layer (B) composed of 0 to 40% by weight of a polyesteramide resin, comprising an aliphatic ester unit and an aliphatic amide unit, and 60 to 100% by weight of an aliphatic polyester resin and on at least one face of a layer (A) composed of 50 to 100% by weight of a polyesteramide resin and 0 to 50% by weight of an aliphatic polyester resin, whereby the thickness of said layer (A) amounts to 50 to 90% of the overall thickness of the film.

19. A multilayer film according to claim 18.

INTERNATIONAL SEARCH REPORT

International Application No
PCT/EP 00/03380

A. CLASSIFICATION OF SUBJECT MATTER
IPC 7 C08K5/00

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)
IPC 7 C08K C08G C08L

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)

WPI Data, PAJ, EPO-Internal

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category °	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	US 5 644 020 A (R. TIMMERMANN ET AL) 1 July 1997 (1997-07-01) column 4, line 22 - line 67; claims	1-4,6,7
X	US 5 216 043 A (D. RUTHERFORD ET AL) 1 June 1993 (1993-06-01) column 4, line 51 - line 63; claims 1,17,19 column 5, line 22 - line 26	1-10
X	WO 99 05207 A (MONSANTO CO) 4 February 1999 (1999-02-04) claims	1-4,6,7

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[X] Further documents are listed in the continuation of box C.

[X] Patent family members are listed in annex.

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Date of the actual completion of the international search	Date of mailing of the international search report
25 July 2000	02/08/2000

Name and mailing address of the ISA	Authorized officer
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INTERNATIONAL SEARCH REPORT

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PCT/EP 00/03380

C.(Continuation) DOCUMENTS CONSIDERED TO BE RELEVANT

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X,P	EP 0 937 743 A (KABUSHIKI KAISHA KOBE SEIKO SHO ET AL.) 25 August 1999 (1999-08-25) page 4, line 40 - line 55; claims; examples	1-10
X	WO 96 21690 A (BASF AG ET AL.) 18 July 1996 (1996-07-18) page 10, line 19 - line 31; claims	1-4,6,7
X	WO 96 21691 A (BASF AG ET AL.) 18 July 1996 (1996-07-18) page 10, line 13 - line 25; claims	1-4,6,7
A	DATABASE WPI Section Ch, Week 199441 Derwent Publications Ltd., London, GB; Class A23, AN 1994-330141 XP002143312 & JP 06 256481 A (TOYOBO KK), 13 September 1994 (1994-09-13) abstract	11-29

INTERNATIONAL SEARCH REPORT

Information on patent family members

International Application No
PCT/EP 00/03380

Patent document cited in search report	Publication date	Patent family member(s)	Publication date
US 5644020 A	01-07-1997	DE 4327024 A	16-02-1995
		EP 0641817 A	08-03-1995
		JP 7102061 A	18-04-1995
US 5216043 A	01-06-1993	AU 3248093 A	19-07-1993
		BR 9206906 A	30-05-1995
		CA 2125572 A	24-06-1993
		DE 69208267 D	21-03-1996
		DE 69208267 T	27-06-1996
		DK 616622 T	24-06-1996
		EP 0616622 A	28-09-1994
		EP 0616570 A	28-09-1994
		ES 2083271 T	01-04-1996
		IL 103713 A	18-06-1996
		JP 7502065 T	02-03-1995
		JP 7502221 T	09-03-1995
		MX 9206972 A	01-06-1993
		PL 170349 B	31-12-1996
		WO 9312171 A	24-06-1993
		WO 9311941 A	24-06-1993
		US 5470526 A	28-11-1995
WO 9905207 A	04-02-1999	AU 8587998 A	16-02-1999
		EP 0996670 A	03-05-2000
EP 0937743 A	25-08-1999	JP 11269365 A	05-10-1999
WO 9621690 A	18-07-1996	DE 19500755 A	18-07-1996
		AU 4484696 A	31-07-1996
		DE 59600383 D	03-09-1998
		EP 0802941 A	29-10-1997
		ES 2119563 T	01-10-1998
		FI 972964 A	11-07-1997
		JP 11500761 T	19-01-1999
		NO 973229 A	11-09-1997
		US 5880220 A	09-03-1999
WO 9621691 A	18-07-1996	DE 19500754 A	18-07-1996
		AU 4390996 A	31-07-1996
		DE 59600362 D	27-08-1998
		EP 0802940 A	29-10-1997
		ES 2119560 T	01-10-1998
		JP 11511767 T	12-10-1999
		US 5863991 A	26-01-1999
JP 6256481 A	13-09-1994	NONE	